The pursuit of rechargeable non-aqueous lithium–oxygen battery cathodes

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Abstract

To satisfy the energy storage needs of society in the long-term, an advance in battery energy density is required. The lithium–oxygen battery is one of the emerging opportunities available for enhanced energy storage. The challenge for the Li–O2 battery is the progress of development of the O2-cathode that allows reversible formation of Li2O2 in a stable electrolyte within its pores.

1. Introduction

A breakthrough in energy density is required to satisfy the energy storage needs of society in the long-term. Lithium–air batteries (hereafter referred to as lithium–O2 batteries since O2 is the reactant) have theoretical specific energies almost 10 times that of the state-of-the-art Li-ion battery technology (Table 1). For practical energy densities these values reduce to factor of 2–3 improvement, which would still provide a major step change in energy storage capability [1]. Certainly if the lithium metal anode would operate successfully in a Li–O2 cell, then the positive energy storage benefit for the Li metal/LiCoO2 cell would also be realised (Table 1). Until recently non-aqueous Li–O2 cells have been in the background of battery science, but since the demonstration of the working principle of a rechargeable Li–O2 cell by Abraham and Jiang [2], and then multiple cycling of a lithium–O2 cell by Bruce and co-workers [3], there has been a “gold rush” by research groups around the world to develop this emerging system for energy storage applications [4–10]. In this fast moving field there have already been a number of reviews on all aspects of the Li–O2 cell [1,11–17]. However, the unique and crucial aspect of the Li–O2 cell is the reaction (or reactions) at the O2-cathode. It is this topic which the present review addresses. Within the limited space available, we cannot hope to review all the excellent work that has taken place on Li–O2; instead we will focus solely on recent developments of the O2 cathode for Li–O2. The review starts with an overview of the non-aqueous Li–O2 cell followed by sections on the role of the electrolyte, the potential role of catalysts and finally on optimisation of porosities and creating novel electrode architectures for air cathodes to increase the power capability of Li–O2.

2. The non-aqueous Li–O2 cell

Unlike a conventional battery where the reagents are contained within the cell, the Li–O2 cell uses oxygen from the atmosphere. The Li–O2 cell can be thought of as a battery–fuel cell hybrid, although it is more a derivative of metal–air batteries (e.g. Zn–air). A schematic representation of the rechargeable non-aqueous Li–O2 cell is shown in Fig 1. On discharge, lithium ions formed at the lithium metal anode are transported across the electrolyte and into the pores of the O2-cathode. O2 from the atmosphere enters the cathode, and dissolves into the electrolyte within the pores. It is then reduced at the porous carbon electrode surface by electrons from the external circuit and combines with Li+ from the electrolyte, leading to the formation of solid Li2O2 as the final discharge product. Somewhat surprisingly, the reaction is reversible, Li2O2 can be oxidised, releasing oxygen gas, thus making this an energy storage device, 2Li + O2 \( \rightarrow \) Li2O2 [3].

The large increase in theoretical specific energy on migrating from Li-ion to Li–O2 arises because Li2O2 in the cathode can store more Li, and hence more charge, than LiCoO2 per unit mass and Li metal stores more charge per unit mass than graphite (C6Li). Some authors have reported that discharge to Li2O is possible [4,6,19], which would further enhance the potential for stored energy (Table 1). However, the oxidation of Li2O may require even greater overpotentials during charge, unless suitable catalysts can be employed [20].

3. Mechanism of ORR and OER in non-aqueous Li–O2 cells

Studies of O2 reduction in non-aqueous electrolytes have been carried out for several decades, but very few addressed the conditions found in Li–O2 cells [21–26]. In this respect, if progress is to be made on Li–O2 batteries, it is important to understand the...
Table 1

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<thead>
<tr>
<th>Battery Description</th>
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<tr>
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*a* Density of LiCoO₂ = 5.16 kg L⁻¹ [18].

*b* Based on the sum of the volumes of Li at the beginning and Li₂O₂ at the end of discharge.

*c* Based on the sum of the volumes of Li at the beginning and Li₂O at the end of discharge.

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The fundamental mechanism of O₂ reduction in the presence of Li⁺ and the formation of Li₂O₂, as well as the mechanism of Li₂O₂ oxidation on charging. Recent detailed electrochemical investigations have probed the influence of salt and solvent on the O₂ reduction reaction (ORR) [4,9,27]. In situ Raman microscopy and mass spectrometry have also been applied [3,28], which offer the important benefit of identifying directly the species involved in the reactions (intermediates and products) on discharge and charge.

The current consensus from the various studies is that the mechanism of O₂ reduction on discharge is:

\[
\text{O}_2 + e^- \rightarrow \text{O}_2^- \quad (1a)
\]

\[
\text{O}_2^- + \text{Li}^+ \rightarrow \text{LiO}_2^- \quad (1b)
\]

\[
2\text{LiO}_2^- \rightarrow \text{Li}_2\text{O}_2 + \text{O}_2 \quad (1c)
\]

However, other studies suggest the occurrence of direct reductions [4,9]:

\[
\text{LiO}_2 + \text{Li}^+ + e^- \rightarrow \text{Li}_2\text{O}_2 \quad (1d)
\]

\[
\text{Li}_2\text{O}_2 + \text{Li}^+ + e^- \rightarrow 2\text{LiO} \quad (1e)
\]

Reaction (1d) occurs at lower voltages than 1(a) [28] and there has to date been incomplete evidence for the formation of Li₂O.

For the oxygen evolution reaction (OER) gas analysis from charging of Li₂O₂ shows that the oxidation of Li₂O₂ follows:

\[
\text{Li}_2\text{O}_2 \rightarrow 2\text{Li}^+ + 2e^- + \text{O}_2 \quad (2)
\]

The reaction course on charging is not the reverse of discharge; the latter involves O₂ as an intermediate, whereas from the evidence provided from the experiments so far performed, the former does not. The strongest confirmation is from the utilisation of propylene carbonate (PC) as the electrolyte [3,28]. As discussed in further detail later, PC is unstable in the presence of O₂ leading to by-products including CO₂ [29]. Because of the absence CO₂ detected during the charging of Li₂O₂ in PC, it can be inferred that no O₂ is generated during the charging process. Furthermore in situ Raman (Fig. 2), shows no signal pertaining to O₂ or LiO₂ during the charging of Li₂O₂ on Au [28].

4. Challenges for the O₂-cathode

The challenges for the O₂ cathode are shown in Fig. 3. A typical O₂-cathode is comprised of a carbon black mixed with a polymeric binder. The porous carbon O₂-cathode is required to ensure a large electrolyte/electrode surface area and accommodate the insoluble discharge product (Li₂O₂), as well as to facilitate oxygen diffusion to the reaction site through the cathode film. In addition, the porous carbon network must provide enough conductivity to deliver electrons to the reaction site efficiently with low overall impedance. A homogenous distribution of a nano-sized catalyst may also be required to maximise the performance by increasing the round-trip efficiency by lowering the voltage gap between charge and discharge processes; however this has been challenged recently as discussed below. For practical Li–O₂ cells an exterior O₂ permeable membrane is required to prevent the ingress of water and carbon dioxide, whilst still allowing the free passage of oxygen. Progress on membrane development is reported in recent reviews [1,17]. Alternatively the Li–O₂ cell could be used without a membrane if O₂ gas is obtained from an air stream scrubbed of H₂O and CO₂. Such engineering solutions would lead to the lowering of possible energy storage of the final system.

5. The electrolyte

The electrolyte is currently the biggest obstacle to progress in Li–O₂ cells. Understanding, controlling and hence eliminating side reactions is a significant undertaking. The electrolyte must be stable to O₂ and its reduced species, as well as the Li₂O₂ compounds that form on discharge; it must exhibit sufficient Li⁺ conductivity, O₂ solubility and diffusion to ensure satisfactory rate capability, as well as wet the electrode surface and possess low volatility to avoid evaporation at the cathode.
5.1. Organic carbonates

Early Li–O2 cells employed an organic carbonate-based electrolyte, either PC or a blend of cyclic and linear carbonates e.g. ethylene carbonate:dimethyl carbonate. Typical lithium salts were lithium hexafluorophosphate (LiPF6) or lithium bis(trifluoromethylsulfonyl)imide (LiTSFI). Organic carbonate-based electrolytes were chosen because of their low volatility, compatibility with Li metal and high oxidation stability (>4.5 V vs. Li/Li+), and a lower toxicity when compared to other organic solvents.

Aurbach et al. [30] indicated organic carbonate instability in the presence of O2 in the early 1990s when studying the effects of contaminants (O2, H2O) on the performance of Li-ion batteries. Nevertheless, many groups reported multiple cycling of Li–O2 cells [2,3,7,31]. It was assumed that the ability to cycle such cells implied reversible formation of Li2O2 while the capacity fading was due to side reactions with the electrolyte.

In 2010 Mizuna et al. [10] and Bruce and co-workers [32] independently reported data which demonstrated that instability in organic carbonate was a much more important issue. Mizuna showed infrared data that revealed the presence of RCOO2Li and Li2CO3 in discharged cells, whilst Bruce and co-workers presented data that showed the gas released on recharge of a discharged cell was predominantly CO2 (98% CO2 vs. 2% O2) [32] (see Fig. 4). Significantly, these studies revealed that there is little or no evidence of Li2O2 formation occurring in parallel with the electrolyte degradation. Further data soon emerged from IBM [8], PNNL [33,34] and ORNL [35] that supported these early studies and ruled out the remarkable ability of Li–O2 cells with organic carbonate electrolytes to sustain cycling (up to 100 cycles) to be due to reversible Li2O2 formation. Instead, the cycling of Li–O2 cells in organic carbonate occurs by degradation of the electrolyte on discharge to form lithium propyl dicarbonate (C3H6(OCO2Li)2), Li2CO3, HCO2Li, CH3CO2Li, CO2 and H2O, with these decomposition products being oxidised on charge, i.e. by repeated cycles of irreversible degradation and not reversible Li2O2 formation [29]. The implication of these findings is that rechargeable Li–O2 cells containing organic carbonate electrolytes will not work. For a rechargeable battery truly reversible reactions must occur at the electrodes.

5.2. Ethers

Attention switched recently to the study of ethers, most notably tetraglyme, (CH3O(CH2CH2O)4CH3) [27,36], triglyme (CH3O(CH2CH2O)3CH3) [37] and dimethoxyethane (DME, CH3OCH2CH2OCH3) [8,27,38,39]. Such ethers are more stable than organic carbonates towards reduced O2 and do exhibit Li2O2 formation at ~2.7 V, at least on the first discharge. Investigation of linear (diglyme, triglyme and tetraglyme) and cyclic (1,3 dioxolane, 2-methyl tetrahydropyran) ethers in Li–O2 cells established that electrolyte degradation does take place leading to the formation of Li2CO3 and Li2O2 [40]. Studies of DME in Li–O2 cells by mass spectrometry showed that only ~60% of the O2 consumed on discharge is released on charge [8]. Oligoether-functionalised silane based electrolytes have recently been reported to show a greater stability to oxygen reduction products and no Li2CO3 was observed in the FTIR of discharged electrodes [41]. Though ethers are more stable than carbonate electrolytes, they are not yet the final solution to the challenge of identifying a suitable non-aqueous electrolyte for Li–O2 batteries that allows multiple cycling with little to no fading [8,40]. As shown by powder XRD (Fig. 5), Li2O2 is present on the 1st discharge but is absent on the 5th cycle discharge.

5.3. Ionic liquids

Hydrophobic ionic liquids have properties that are favourable for practical Li–O2 cells as they are non-flammable, non-volatile, and, due to their hydrophobic properties, will better protect the metal anode from moisture in comparison to other aprotic...
Solvents. However, O₂ solubility and diffusivity are likely to be lower in this class of electrolytes, which is likely to impact on the power output of such cells. Superoxide formation has been demonstrated in several ionic liquids using cyclic voltammetry without the presence of a metal salt [42–45]. In the context of the Li–O₂ cell, two recently published papers show the effect of a lithium salt on the voltammogram. Both papers show that the presence of Li⁺ leads to superoxide instability with the formation of insoluble lithium–oxygen species that passivate the glassy carbon working electrode. In addition Allen et al. [46] reported that, conversely a gold electrode demonstrated more efficient recharging, with multiple cycling without passivation, providing evidence that the role of the electrode.Much attention has been devoted to the stability or instability of the solvent; fewer studies exist on understanding the role of the lithium salt. XPS data [52] have shown that LiPF₆ exposure to Li₂O₂ causes the formation of a thick layer of LiF on the Li₂O₂. LiPF₆ decomposition has also been observed by XPS on MnO₂/C O₂−cathodes from discharged Li–O₂ cells [35]. This is deleterious to the Li–O₂ cell both from the loss of lithium and electrolyte salt in the formation of inactive decomposition products, but also from the potential detrimental effect on the oxidation of Li₂O₂ by the modification of the interface between Li₂O₂ and the O₂−cathode surface. Decomposition products of lithium bis(oxalate)borate (LiBOB) have also been reported, both by chemical reaction with Li₂O₂ [52] or LiO₂ [53]. LiBOB has been shown to be more stable against Li₂O₂ [52], but may suffer from lower oxidative stability whilst LiTFSl and lithium bis((fluorosulfonyl)amide (LiFSI) have been proposed as more stable salts for use in the Li–O₂ cell [53].

5.6. Electrolyte additives

One method of improving the performance of Li–O₂ cells is to use additives that increase the solubility of Li₂O₂ in the electrolyte. This could increase the oxidation kinetics and hence charging rate. Additives that increase the O₂ solubility could increase the discharge rate. Enhancing the power capability of Li–O₂ cells is an important target. Organoboron Lewis acids such as tris(pentafluorophenyl) borane (TPFPB) B(C₆F₅)₃ have been reported to increase of the dissolution of Li₂O₂ in carbonate-based organic solvents [54]. Boron esters have also been investigated [55] and the presence of diol-monoacid-borate was shown to increase the solubility of Li₂O₂ in EC:DMC by an order of magnitude from 0.29 to 3.2 mmol L⁻¹. Tetrabutyl ammonium cation (TBA⁺) has also been proposed to aid in the dissolution of oxygen reduction products [4]. To increase the solubility of O₂ in electrolytes a small amount (0.5%) of perfluorotributylamine was added to 1 M LiPF₆ in PC [56]. A factor of 5 increase in the peak current for O₂ reduction was recorded. This general research direction looks promising and will need to be repeated in more stable electrolytes, in order to observe and understand the effect of these perfluorinated additives in Li–O₂ cells. Nevertheless, if Li₂O₂ solubility becomes too great, then significant amounts will ultimately migrate as solubilised peroxide to the lithium anode. Insulating films of lithium oxides would then result on the lithium surface, which would detrimental to the functioning of the device by increasing resistivity of the anode interface.

5.7. Binder

In terms of finding stable components for Li–O₂ cells, such as solvent and salt, it should be mentioned that very recent work by Black
et al. [57] have shown that stable binders are also required. They found that superoxide actively dehydrofluorinates polyvinylidene difluoride (PVDF, also known as Kynar-Flex), which is the most commonly utilised binder in O\textsubscript{2}/C\textsubscript{2} cathodes. The resultant by-products react further with the transition metal–oxide catalyst, such as \(\alpha\)-MnO\textsubscript{2}, to produce LiOH which accumulates at the interface, as detected by FTIR and XRD in Li–O\textsubscript{2} studies in ether-based electrolytes [40,57]. Lithiated Nafion has been found to be relatively unreactive with respect to superoxide and therefore may be a preferred future choice for a more stable binder in future Li–O\textsubscript{2} studies [5,57].

5.8. Future electrolytes

The search for an electrolyte that is stable towards reduced oxygen species and Li\textsubscript{2}O\textsubscript{2}, thus permits highly reversible Li\textsubscript{2}O\textsubscript{2} formation/decomposition remains one of the major challenges for the rechargeable Li–O\textsubscript{2} battery. Note that it is not enough to simply demonstrate reversible formation of surface layers of Li\textsubscript{2}O\textsubscript{2}. Although this is useful for mechanistic studies [4,28], it is necessary to show that highly reversible Li\textsubscript{2}O\textsubscript{2} formation/decomposition with a relatively high capacity is possible, if the non-aqueous rechargeable Li–O\textsubscript{2} battery is ever to be possible. Computational methods of screening solvent susceptibility of nucleophilic attack by superoxide have been undertaken and are likely to improve our understanding of suitable candidate electrolytes [58]. Experimentally it is important to use techniques such as FTIR and differential electrochemical mass spectrometry to identify and quantify side reactions and reversibility.

6. The O\textsubscript{2}-cathode

6.1. Substrate effects

The role of the substrate (in most cases carbon) and the catalyst, included to reduce the voltage gap by promoting oxidation of Li\textsubscript{2}O\textsubscript{2}, will become an increasing focus of attention, especially once more stable electrolytes are identified. In particular, understanding how the substrate affects the reaction mechanism, the degree of side reactions and reversibility of Li\textsubscript{2}O\textsubscript{2} formation/decomposition will be crucial areas for study.

Experiments so far have revealed little mechanistic understanding of the role of carbon in the O\textsubscript{2}-cathode. Preliminary modelling studies [59] conclude that carbons with a high concentration of oxygen surface groups promote oxygen reduction. McCloskey et al. [8] found that although electrolyte decomposition was the major source of CO\textsubscript{2} during cell charging, cathode carbon oxidation contributed about 4% of total CO\textsubscript{2} production. Whether the oxidation occurred from carbon surface functional groups directly, or from carbon surface functional groups that had reacted with superoxide to form surface decomposition products, remains to be identified. Furthermore, whether oxygen functional groups on the carbon surface are regenerated by exposure of oxygen reduction products during discharge is another avenue of prospective research in understanding the role of the substrate. Certainly research efforts should be made to ascertain if carbon is an intrinsically suitable conductive substrate for Li–O\textsubscript{2}. Indeed, as mentioned previously, it has been observed that gold is a more reversible substrate than glassy carbon in cyclic voltammetry studies in ionic liquids [46]. Both electrolyte and substrate are important factors to understand and control in the Li–O\textsubscript{2} battery.

6.2. Catalysts for Li–O\textsubscript{2}

The requirement for a catalyst in the Li–O\textsubscript{2} cell is still unclear. Many early studies were carried out in electrolytes containing organic carbonates [3,5,7,19,60,61], which decompose on discharge [10,29]. As a consequence, the catalyst therefore is catalysing the electrolyte decomposition on discharge and the oxidation of the decomposition products on charge [29]. Future studies will have to be repeated in more stable electrolytes, which at least for the first discharge–charge cycle exhibit significant formation and decomposition of Li\textsubscript{2}O\textsubscript{2}. For the OER the effect of various catalysts...
on the electrochemical oxidation of Li₂O₂ (Li₂O₂(s) → 2Li + O₂(g)) have been investigated by charging electrodes constructed with Li₂O₂ included within the as-prepared cathode [62]. PC based electrolyte was employed but this is stable on oxidation so does allow examination of Li₂O₂ oxidation. The study included a variety of transition metal oxides, with nanowires of MnO₂ giving the lowest charging voltage in Li–O₂ cells of 3.5 V vs. Li/Li⁺. Overall, it was demonstrated that catalysts do indeed lower the charging voltage plateau of Li₂O₂ [62].

Conversely IBM [63] recently reported that an OER catalyst is not required due to the observation that oxygen is primarily evolved at low depths of discharge at a potential of 2.9 V, which is only just above the open circuit potential of the discharged cell (2.8 V). Additional work is therefore required to see if O₂ evolution also occurs at low potentials in O₂-cathodes preloaded with Li₂O₂, which will avoid any side-reaction complications from discharge. For the ORR it has been shown that the inclusion of MnO₂ [40] or Pt [63] increase the proportion of electrolyte decomposition in the more stable ether-based electrolytes. The major mechanism leading to increased electrolyte decomposition results from breakdown products, generated by the reaction of superoxide with PVdF binder, reacting with catalysts in the O₂-cathode to form LiOH, as was discussed earlier in the review [57] Lu et al. [64] demonstrated that O₂ adsorption energy on the surface can greatly influence Li⁺-ORR activities and that a volcano dependence can be inferred (Fig. 7). Activities were shown to be in the order of Pd > Pt > Ru ≈ Au > GC. Accompanying work comparing the performance of noble metals with transition metal oxides would be attractive.

6.3. Structure

As shown by Fig. 8, increasing the discharge current drastically reduces the capacity in Li–O₂ cells, indicating the problem with kinetics [11]. This has motivated many authors to examine novel O₂-cathode architectures, in order to increase power capability. In aqueous systems gas diffusion electrodes are employed in which the gas is carried in hydrophilic channels that are in very close proximity. In this way the O₂ is transported mainly in the gas phase, rather than the much slower diffusion in the liquid. Of course such architecture is harder to engineer with non-aqueous electrolytes. Assuming a suitable gas diffusion electrode that ensures facile O₂ transport can be constructed, and then a high electrode surface area is necessary. However high surface areas imply small pores that could become blocked by solid Li₂O₂, so a compromise is necessary between high active surface area and adequate pore size, the compromise is expected to be found in the mesoporous region. Additionally, the optimisation of the relative amounts of carbon, catalyst and binder, and their distribution will be important.

Growing carbon nanofibre carpets onto a porous ceramic substrate produced not only high specific energies of up to 2500 W h kg⁻¹ (based on electrode mass), but also specific power up to 1000 W h kg⁻¹ at a specific energy of 1000 W h kg⁻¹ [38]. Co₃O₄ nanorods grown onto nickel foams have been shown to deliver good performance (up to 4000 A h kg⁻¹ total electrode mass) and low voltage gaps (0.5 V at a low current density of 0.02 mA cm⁻²), but without sustained cycling [65]. Another design solution involved using hierarchical porous graphene [37], where its structure permitted access of O₂ to most of the graphene sheets. Extremely large capacities were reported of up to 15,000 A h kg⁻¹ (based on mass of carbon). On the other hand, increasing the amount of binder with respect to carbon was found to substantially reduce capacities because of pore blockage [66]. Once again when more stable electrolytes are available the ability of these novel structures to sustain many cycles of significant formation and oxidation of Li₂O₂, will be able to be better understood.

7. Summary

The non-aqueous rechargeable Li–O₂ cell holds significant promise for high energy storage applications, but it is a long way from being a technological product. Fundamental challenges in all aspects of the cell, anode, electrolyte and cathode, need to be addressed. The reaction at the cathode (reversible formation of Li₂O₂) is what defines the non-aqueous Li–O₂ cell and represents one of the major current challenges; therefore we have concentrated on this in the present review. Recent studies have shown that finding an electrolyte/electrode combination that permits highly reversible formation/decomposition of Li₂O₂ on sustained cycling (i.e. formation of high purity Li₂O₂ with minimal side reaction on discharge and then its complete oxidation on charge), is a key problem. Rechargeable batteries need to be based on truly reversible reactions and the ability to cycle a cell is not proof of true reversibility. This highlights the importance of using analytical techniques such as FTIR and DEMS, in order to demonstrate the purity for Li₂O₂ formation and its complete oxidation on charge. Major gaps in fundamental knowledge of the Li–O₂ cell remain, for example the true role of the catalyst. As a result, much ground-breaking and exciting science remains to be done.
One fact is certain, society requires energy storage devices with much higher levels of energy storage than ever before. Non-aqueous Li–O2 cells are amongst the few contenders that can exceed the stored energy of Li-ion therefore continuing research efforts are essential in order to solve the scientific challenges of the Li–O2 cell.

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